Micro-structural changes and helium release in thin films of $Er(D,T)_2$

Hydrogen and Helium Isotopes in Materials

February, 2007

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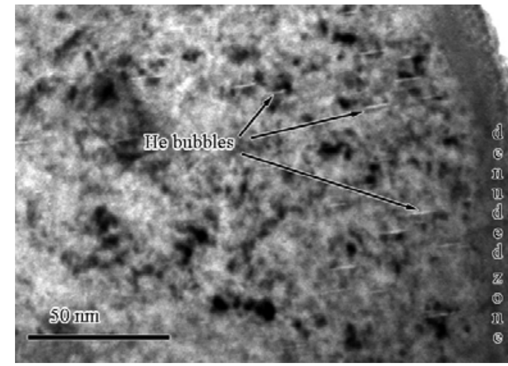
SNL/NM

Dept. 2725

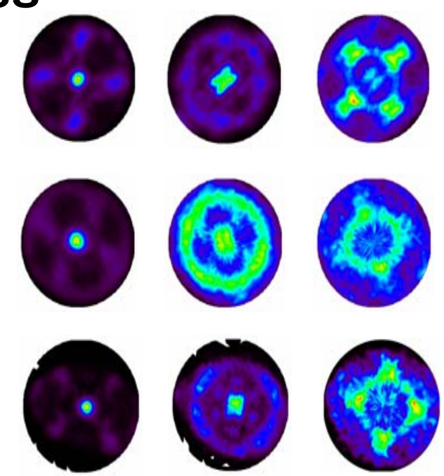
Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company,
for the United States Department of Energy's National Nuclear Security Administration
under contract DE-AC04-94AL85000.

What do we want to learn from this study?

- Changes in micro-structure of films with age
  - $Er(DT)_{2+x}He_x$ – what changes are caused by the decay of tritium?
  - How is the helium stored/released in the film?



- Changes in micro-structure with film thickness
  - Deposition thickness can change by 10% depending on which “ring” of evaporator.
  - Can we learn where helium is being released from?



How the samples were prepared and stored.

- Samples were evaporated by e-beam vapor deposition.
  - Molybdenum substrate.
  - Substrate temperature 450C.
  - Deposition rate 10Å/s.
  - 5000Å, 4000Å, 3000Å, 2000Å, and 1000Å thick Er films.
  - Hydrided after bringing to elevated temperatures.
  - Not all films usable, probably because of substrate contamination.
- Films immediately made into neutron tubes – high vacuum ceramic envelopes.
  - UHV conditions in envelopes.
  - These neutron tubes were not operated.

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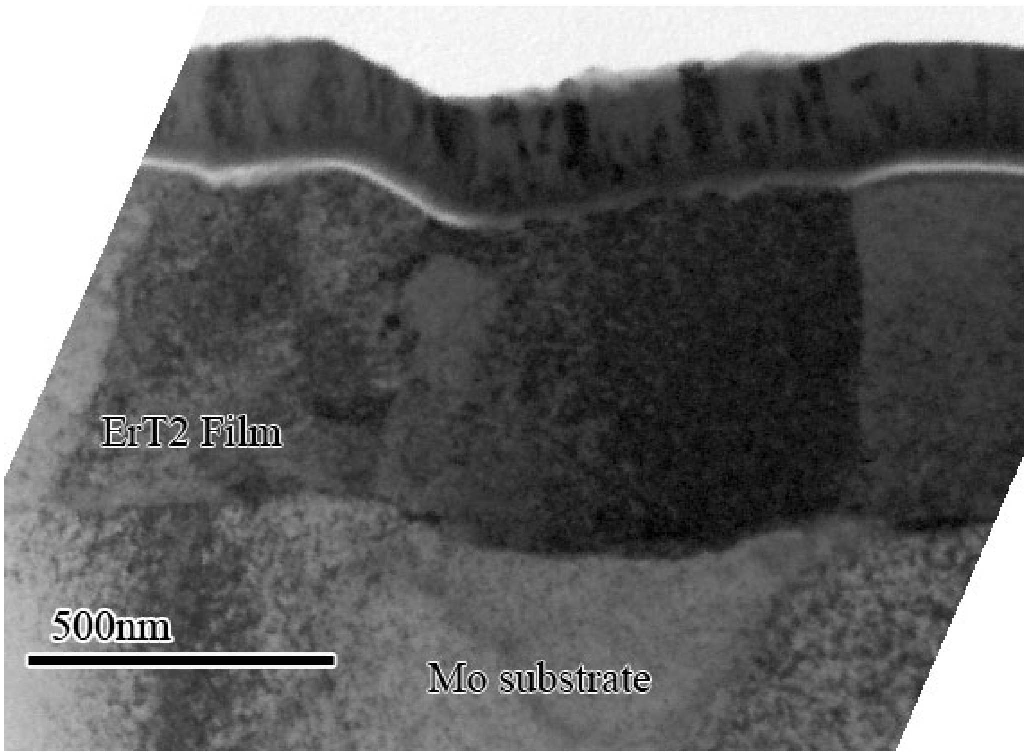
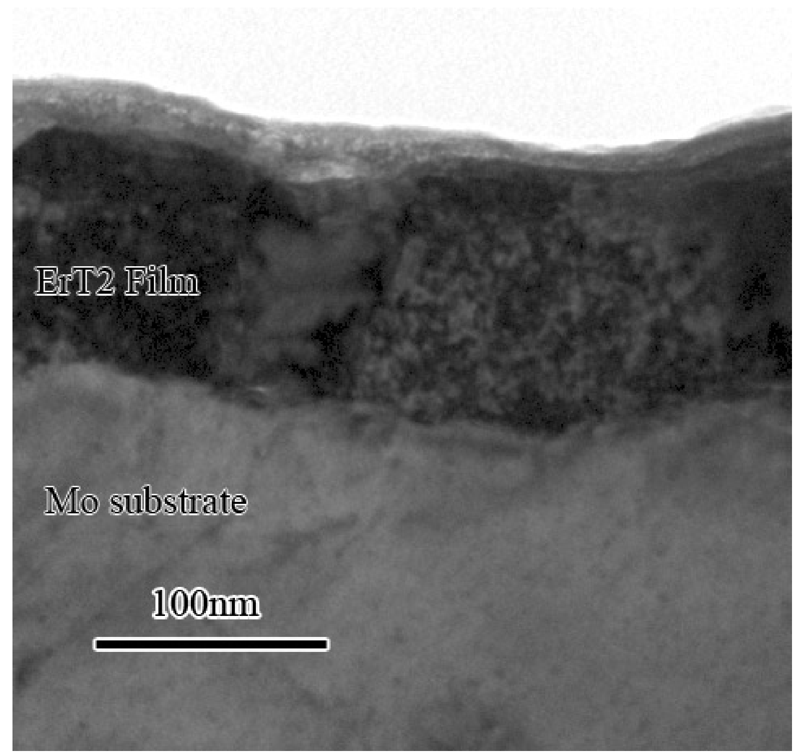
TEM images show that the film has a columnar structure

- **Films of all thicknesses show similar columnar grains.**

5000Å

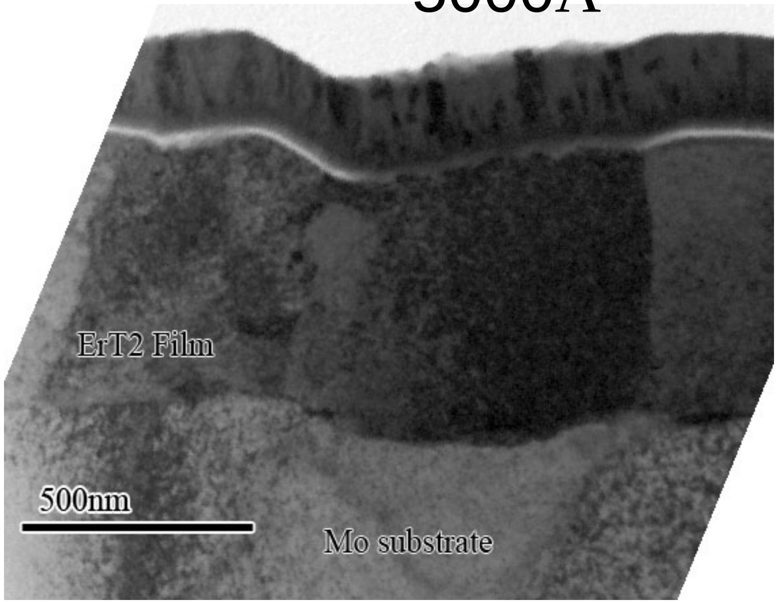


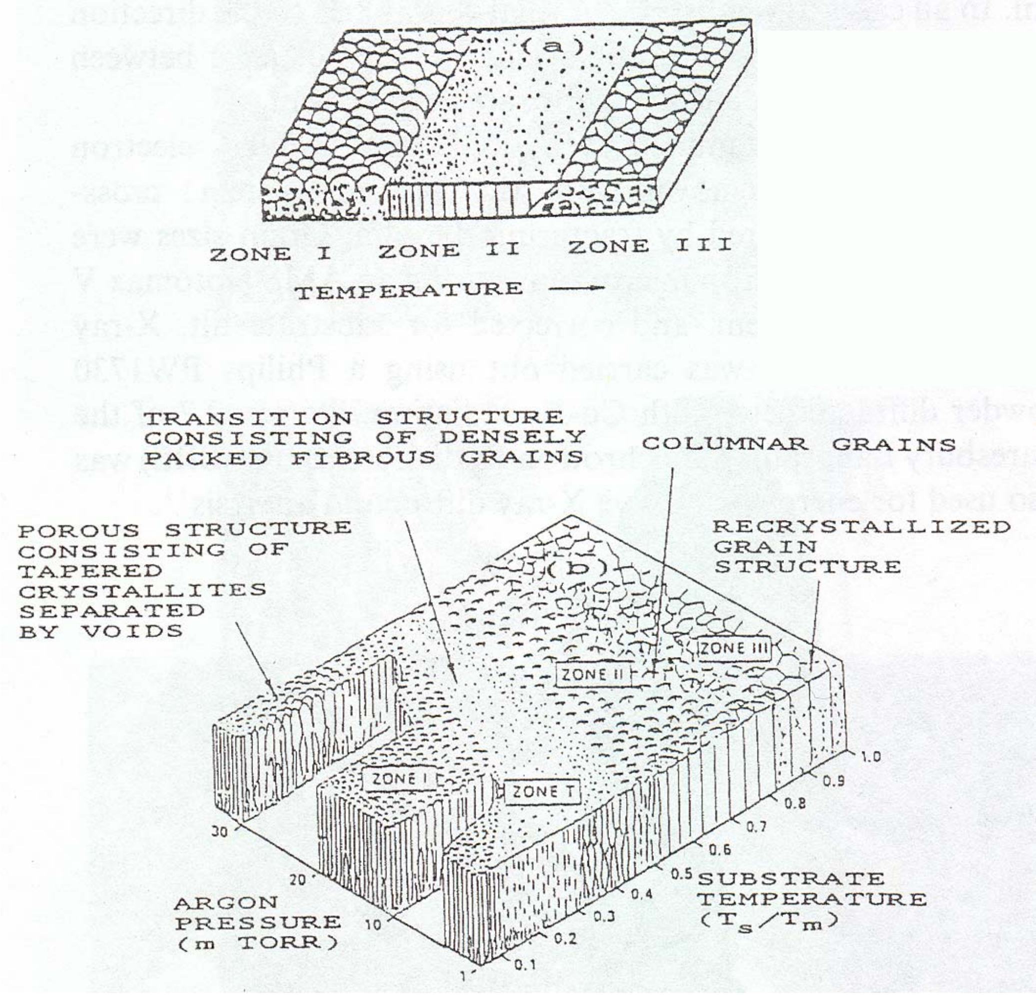
1000Å



The columnar grains fit the growth model of Savaloni and Player.

5000Å

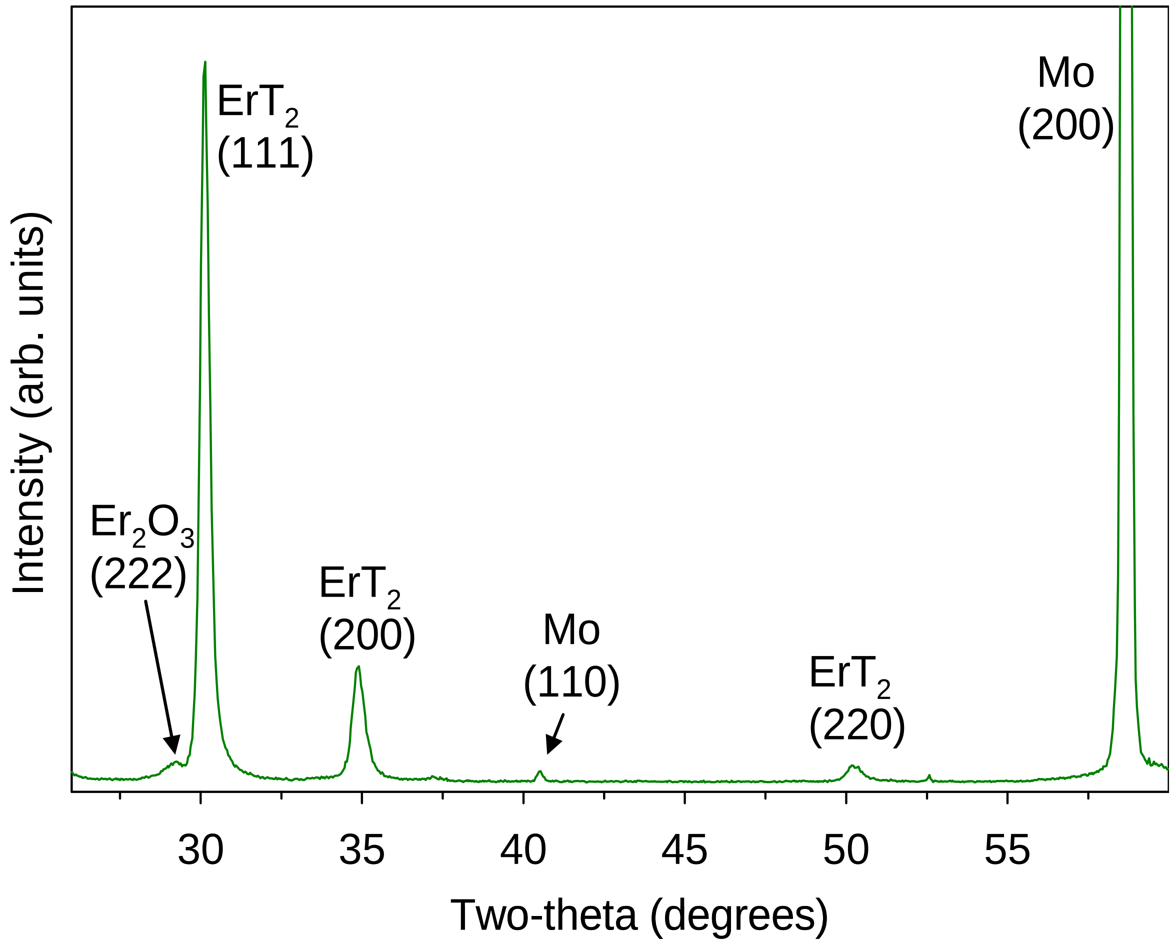




Savaloni et al., Vacuum Vol. 43, pp 965-79 (1992).

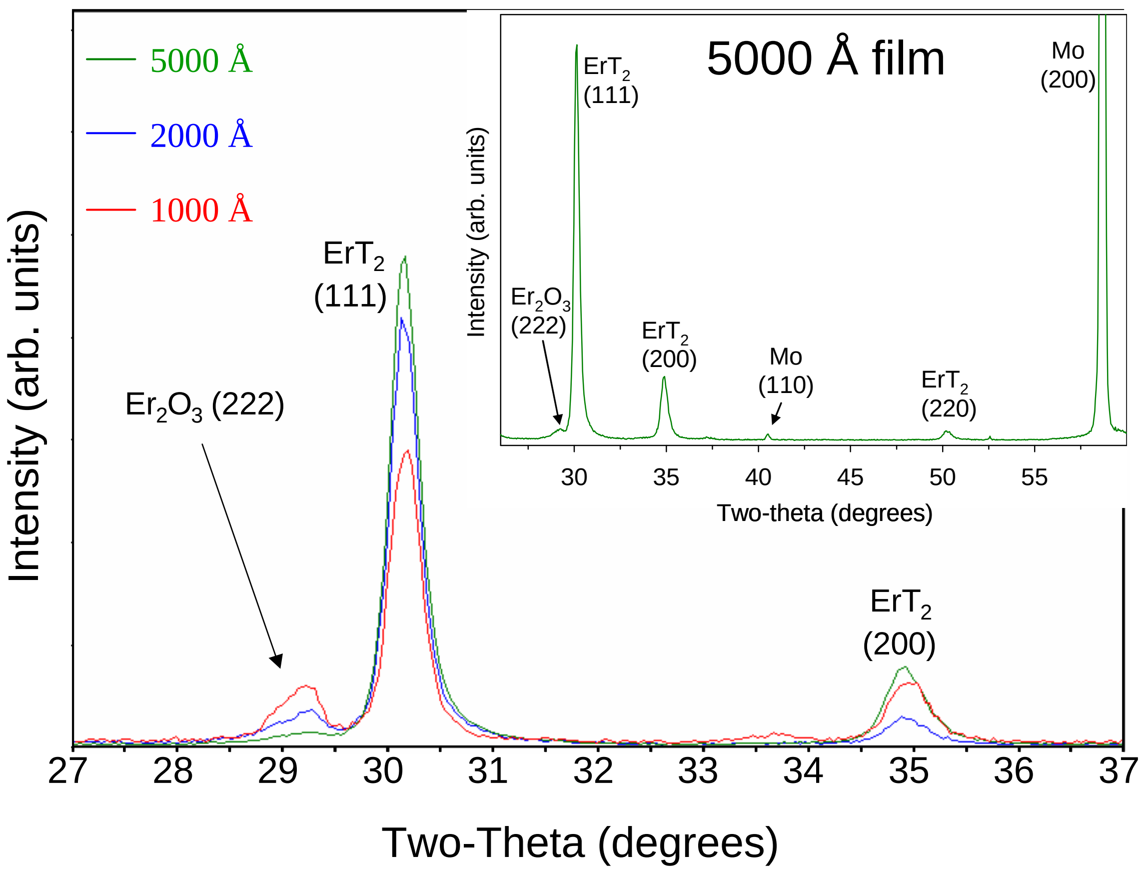
Changes in film texture with film thickness.

- **XRD 2-theta scan for 5000Å $Er(DT)_2$ film.**

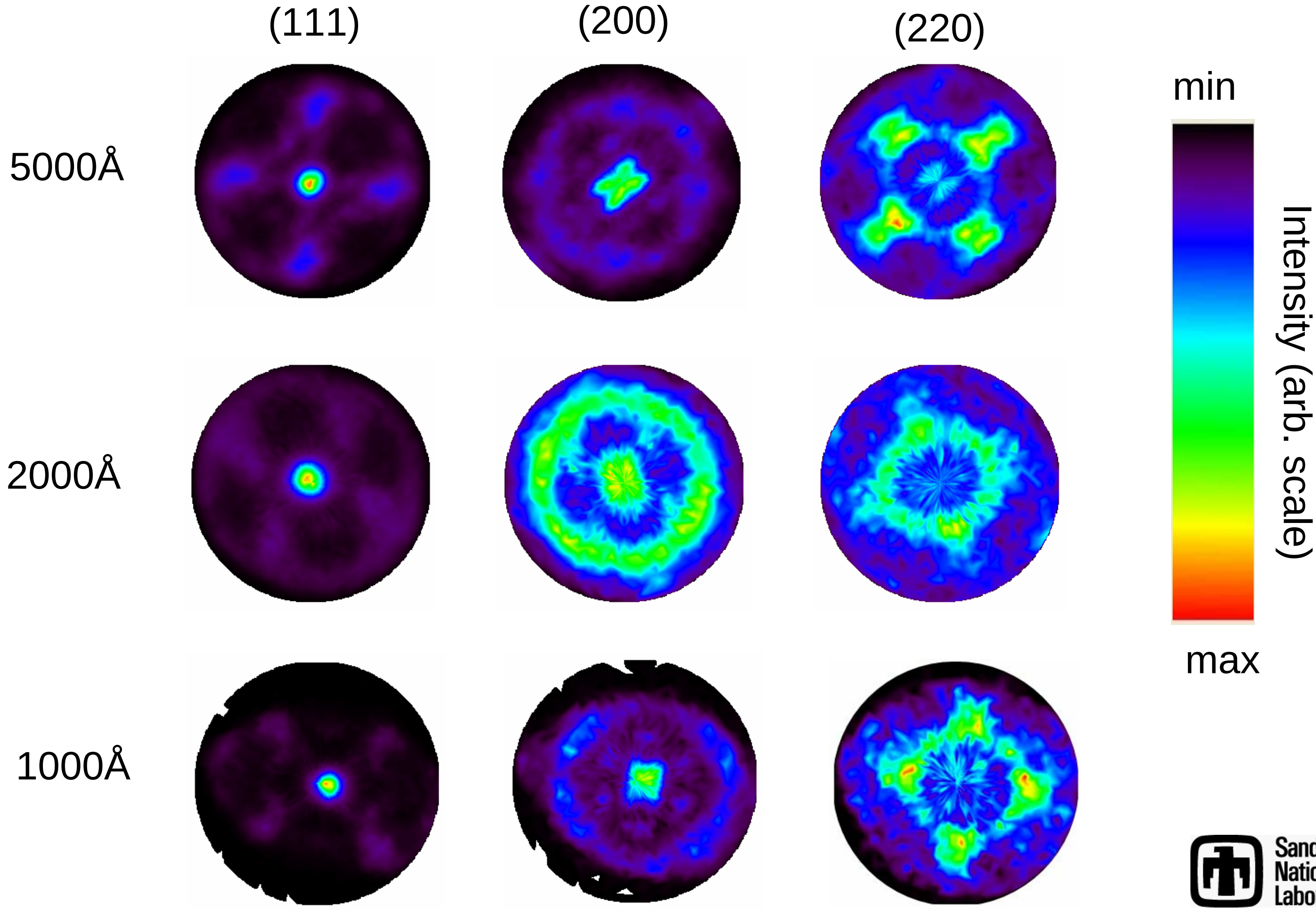


Close up view of the oxide and ErH_2 diffraction lines as a function of film thickness.

- **Texture changes with film thickness**
  - **The thicker the film shows a stronger (111) in plane texture.**
- **The thinner the film, the less oxide.**

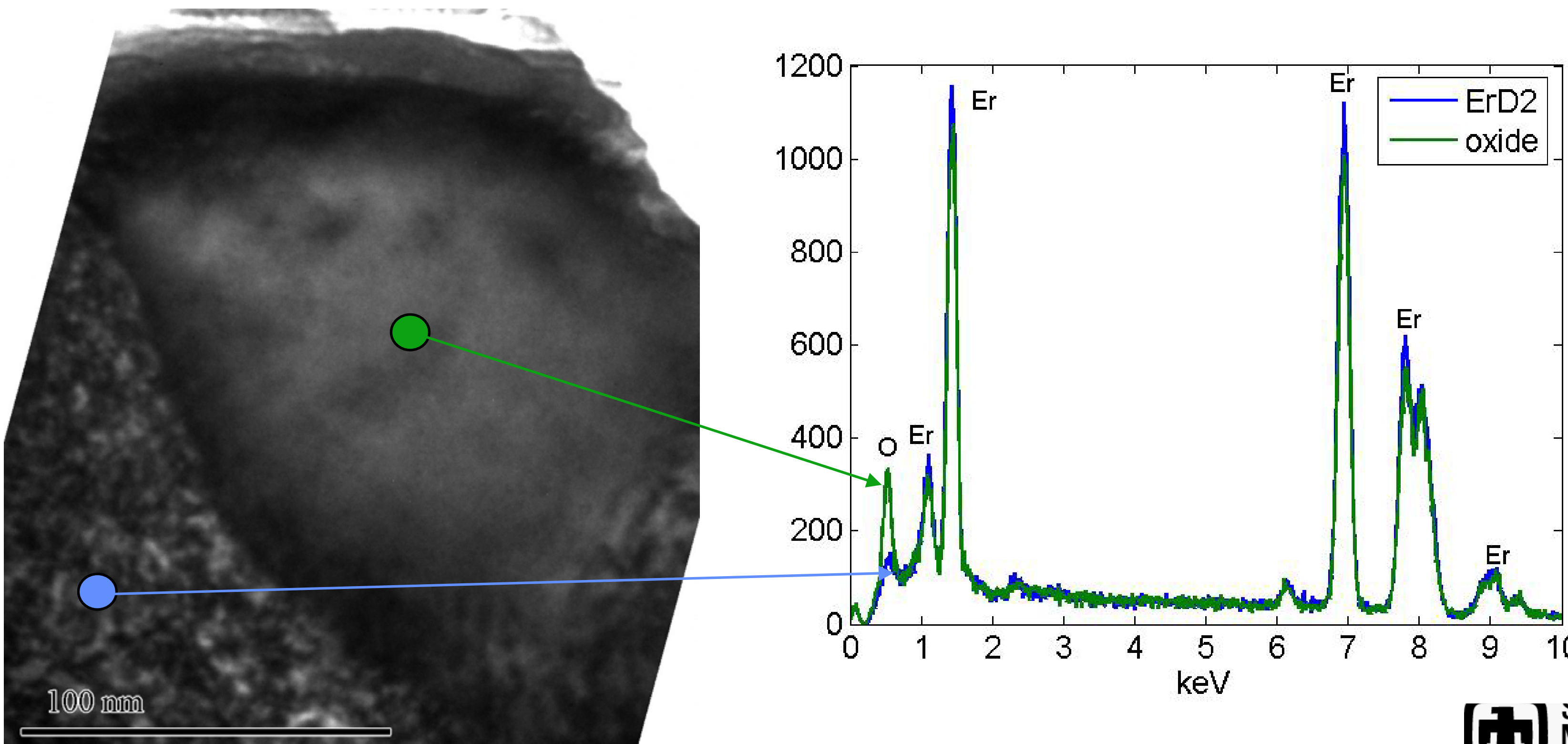


Pole figures show the change in film texture with film thickness.



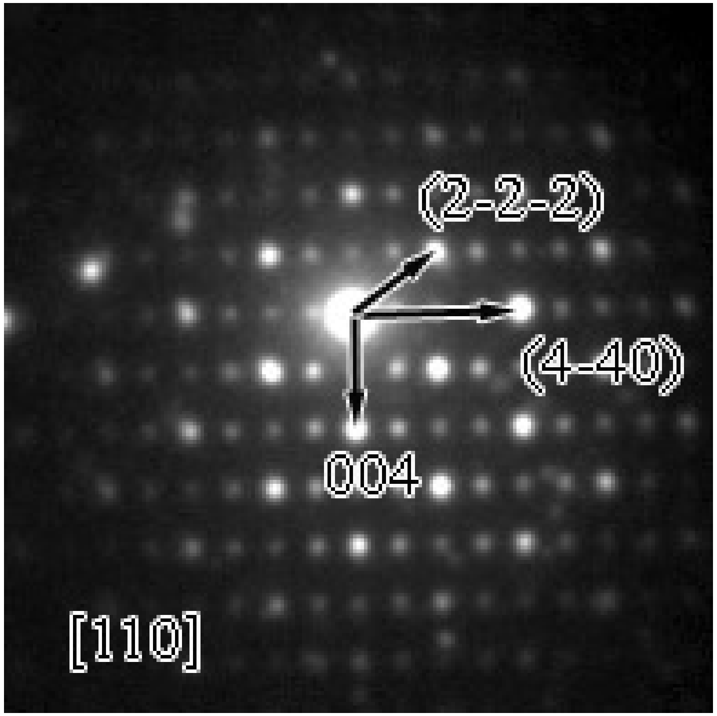
TEM shows very large oxide inclusions

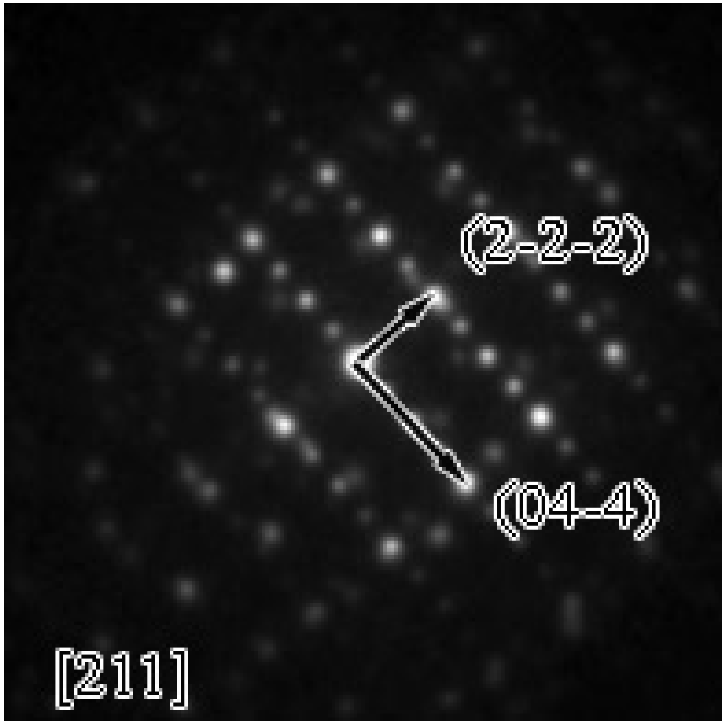
- **All films show large oxide inclusions that often span the film thickness.**



Not only is the inclusion oxygen rich but it has the crystal structure of Er_2O_3

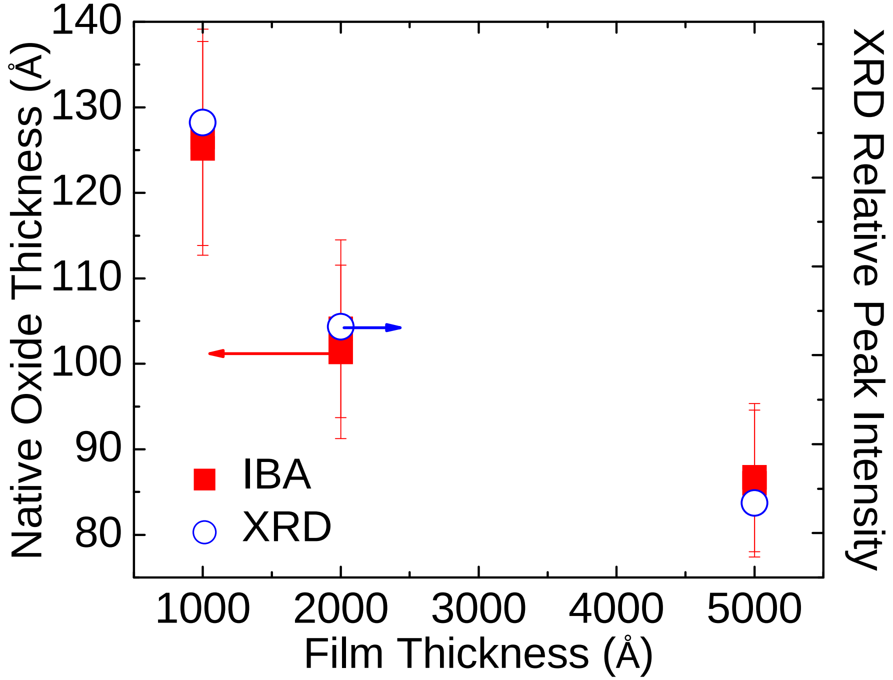
- Diffraction spots index well to the crystal structure of Er_2O_3

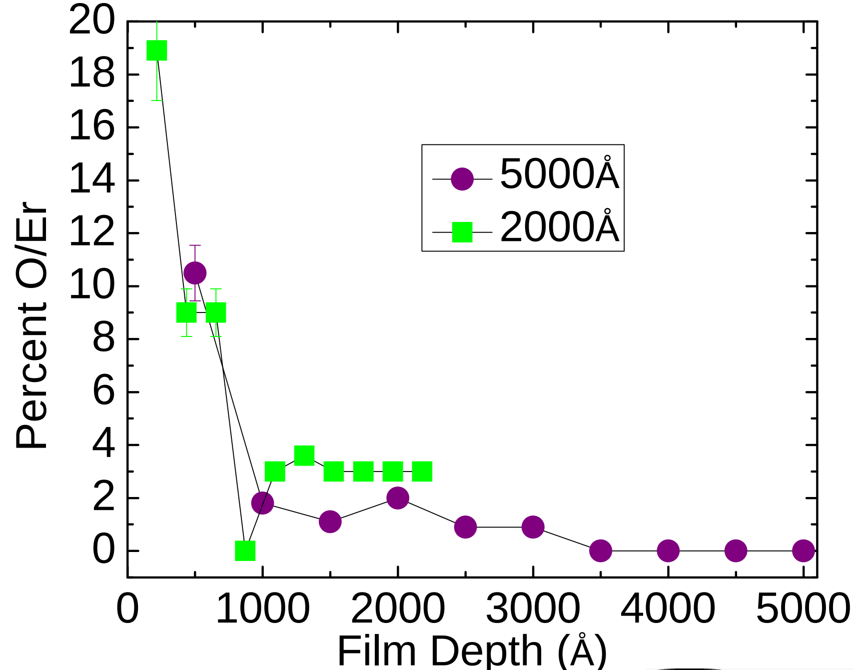




The oxide surface layer also varies with film thickness.

- Oxide layer was measured by IBA.
- Correlated to peak intensity of XRD.
- Bulk oxygen, determined by IBA, similar in all samples.





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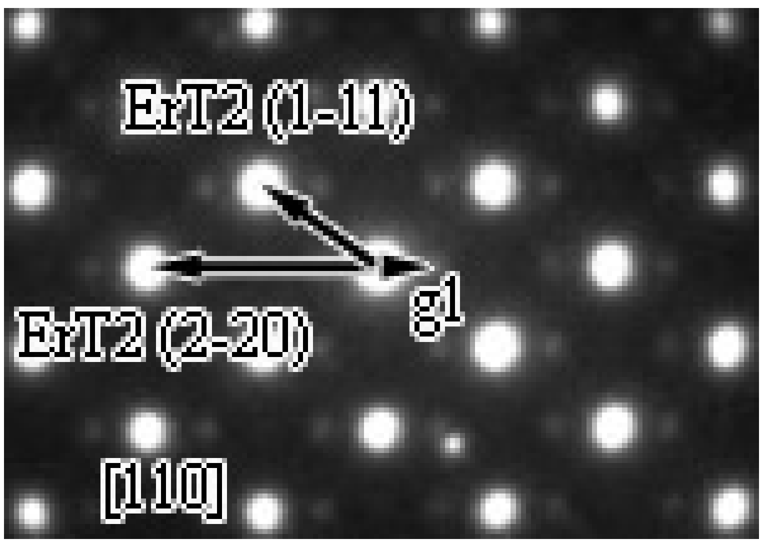
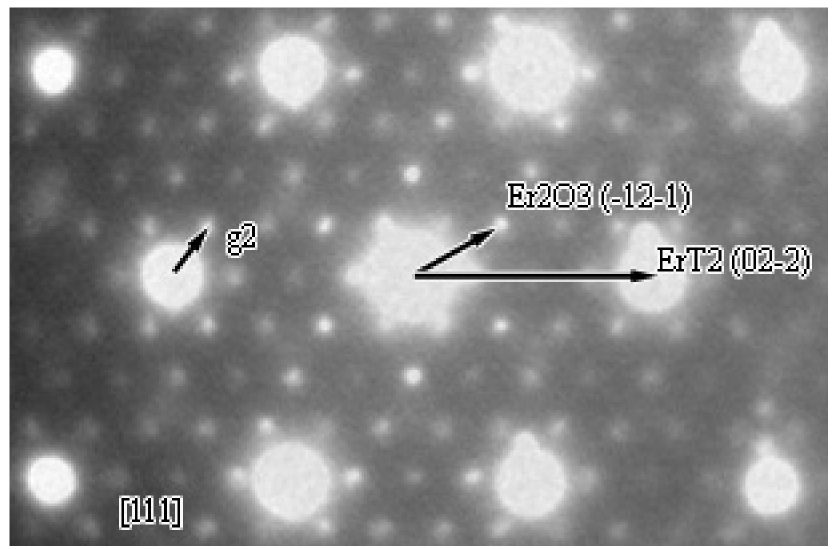
What is the origin of the mysterious secondary spot pattern seen in TEM?

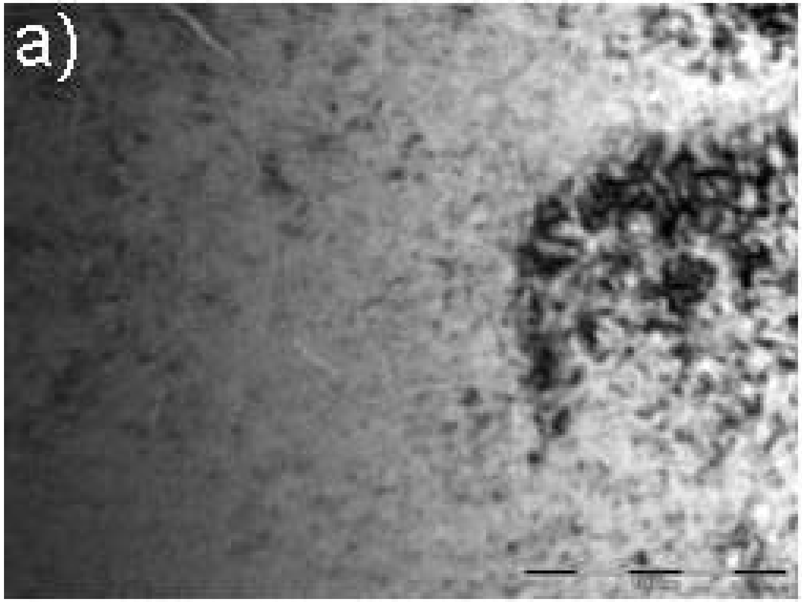
- Secondary spot pattern seen in all of the films.
  - g1 and g2 *d*-spacings are 7.3Å and 7.4Å respectively.

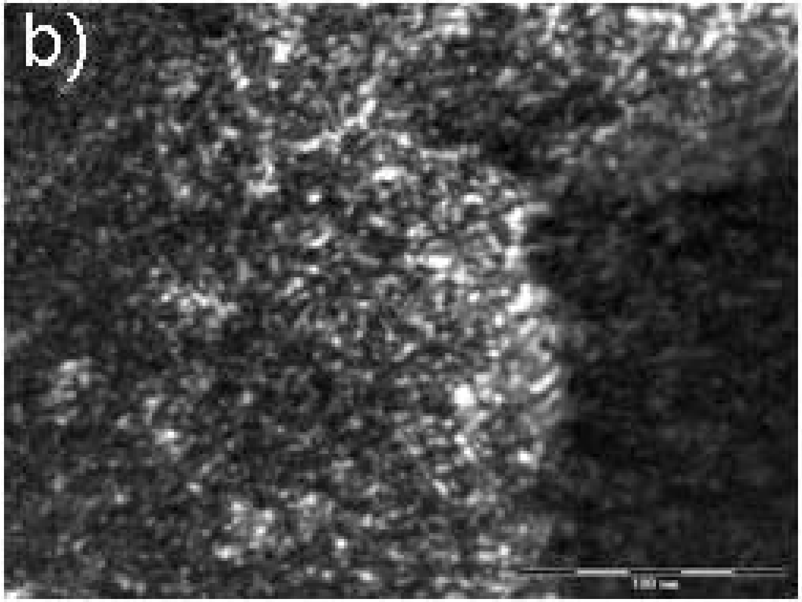




Bright field/Dark field image pairs corresponding to the contrast from the secondary spots

- Bright field/Dark field image pairs.
- 5-10 nm objects dispersed throughout film.





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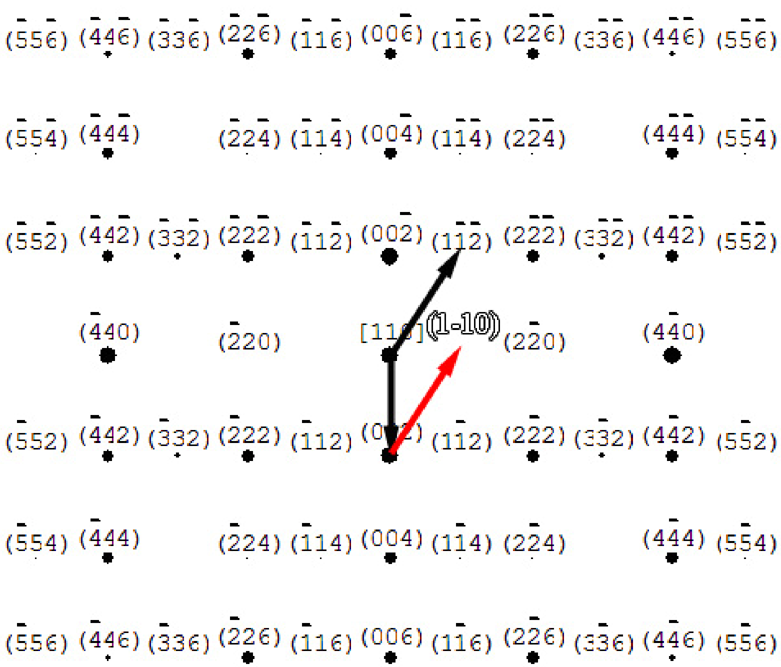
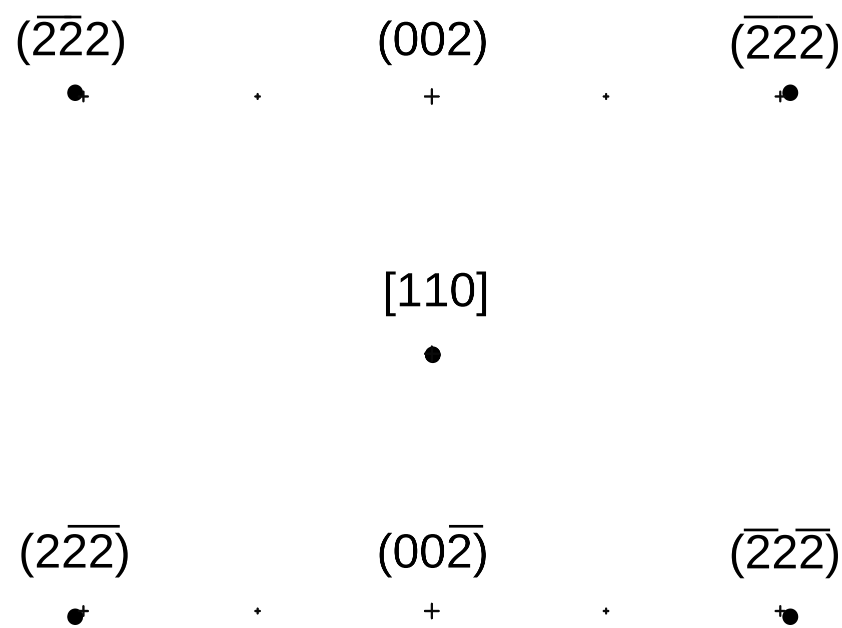
The secondary spots index closely to ErH_2 and Er_2O_3

g	ErH_2 {hkl}/d(Å)	ErH_3 {hkl}/d(Å)	Er_2O_3 {hkl}/d(Å)	Er {hkl}/d(Å)	$Er(OH)_3$ {hkl}/d(Å)	ErO(OH) {hkl}/d(Å)
7.3Å	{100}/5.15	{110}/5.41	{200}/5.27	{100}/3.08	{100}/5.41	{100}/5.62
7.4Å	{110}/3.64	{002}/3.26	{110}/7.45	{002}/2.80	{110}/3.12	{001}/4.06
	{111}/2.97		{222}/3.04			
			{220}/3.73			
	2*{110}/7.28					

Calculated SAED patterns for zone axis of Er_2O_3 show the ErH_2 overlap

- **Red arrow shows double diffraction between (002) and (1-1-2) which results in appearance of (1-10).**





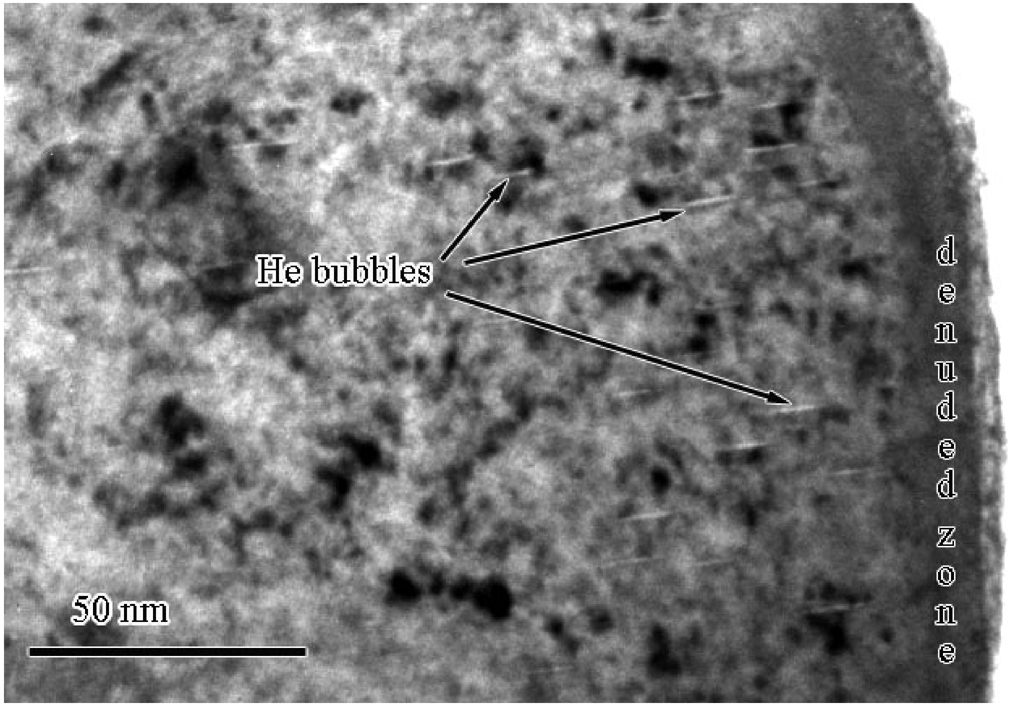
A summary of what we have learned about the “real” film micro-structure

- The films are textured – (111) out-of-plane
- The films have columnar grains.
- The films have large oxide particles.
- The films have an even dispersal of 5-10 nm oxide particles.

All of these films exhibit lenticular helium bubbles

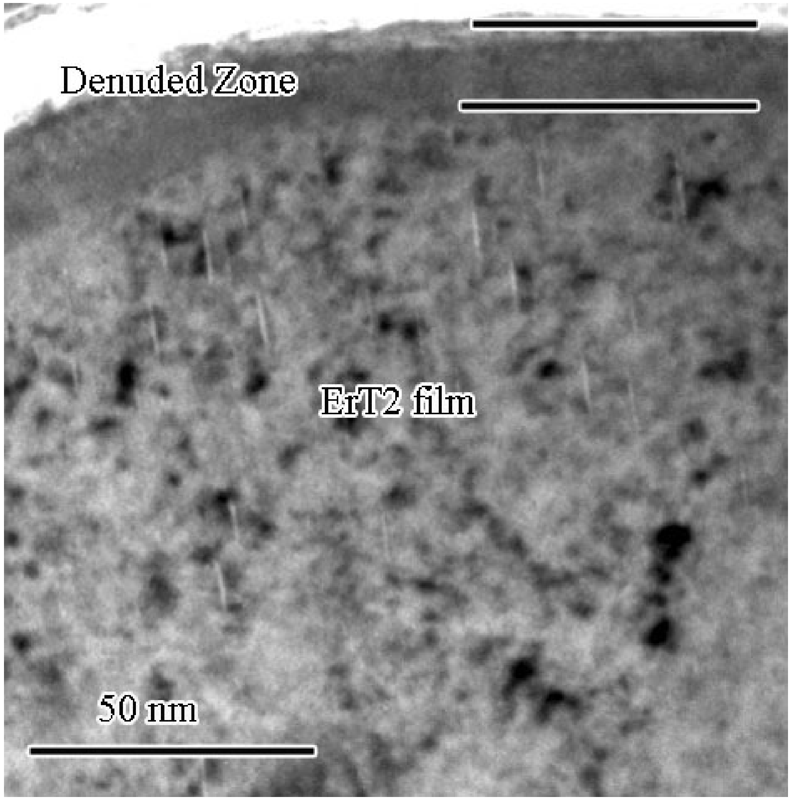
- **Note lenticular bubbles and "denuded zone" or "bubble free zone".**
- **All films show an average bubble length of ~10nm with a bubble volume density of ~5x10^{17}/cm^{3}**

5000Å

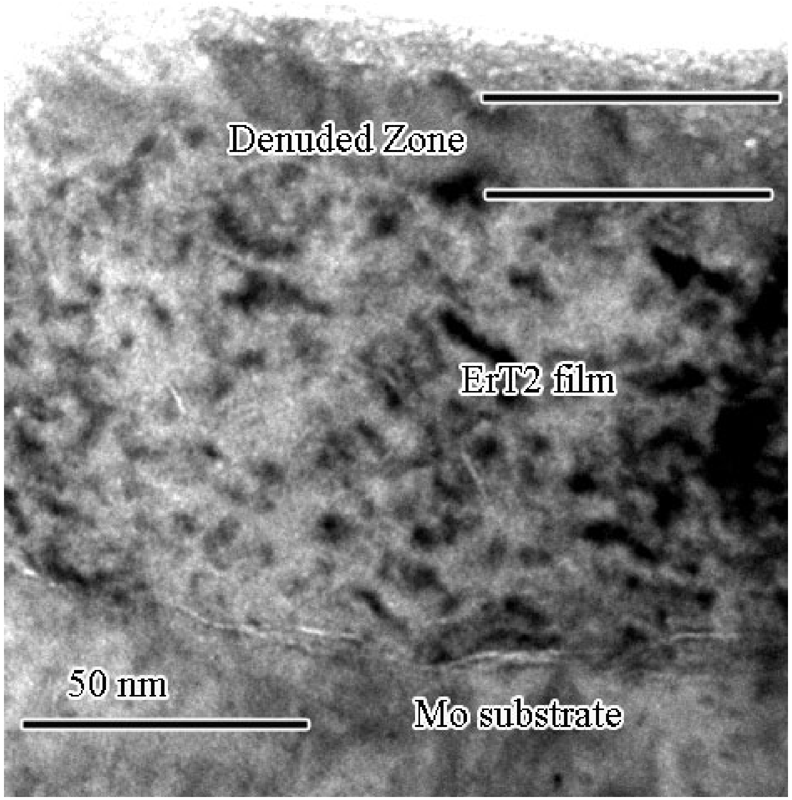


The “denuded zone” is constant.

- **“denuded zone” is very constant at ~15nm for every sample in this study.**



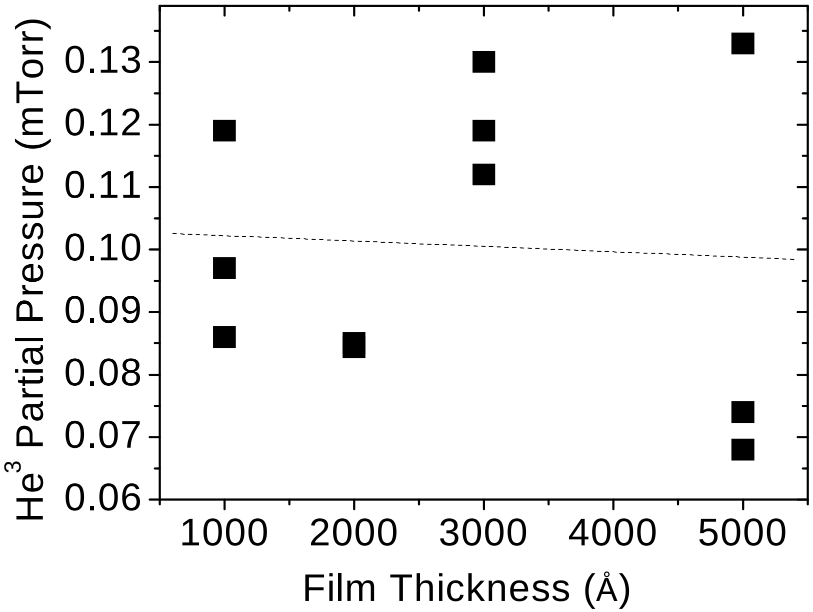
5000Å

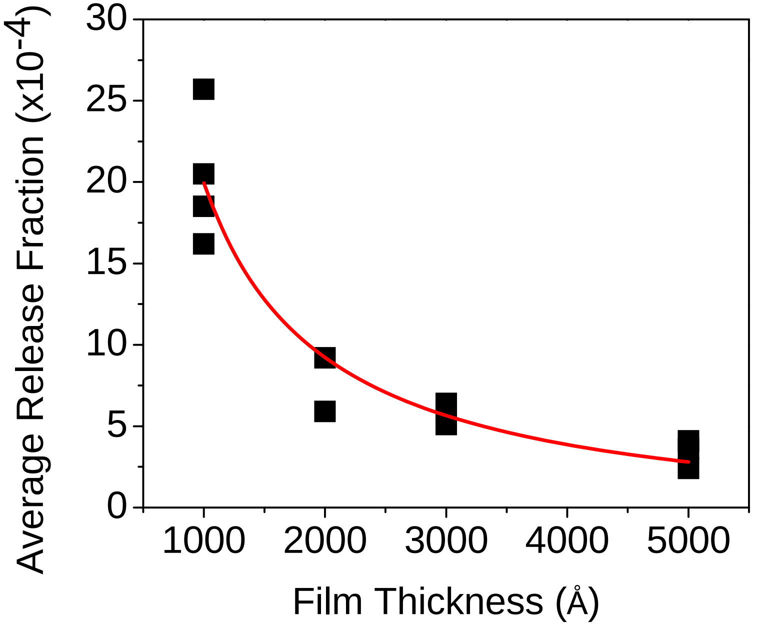


1000Å

How does the helium release amount vary with film thickness?

- **Total amount of helium released is fairly constant.**
- **ARF=(amt. He released)/(amt. He generated)**
- **ARF varies inversely with time.**





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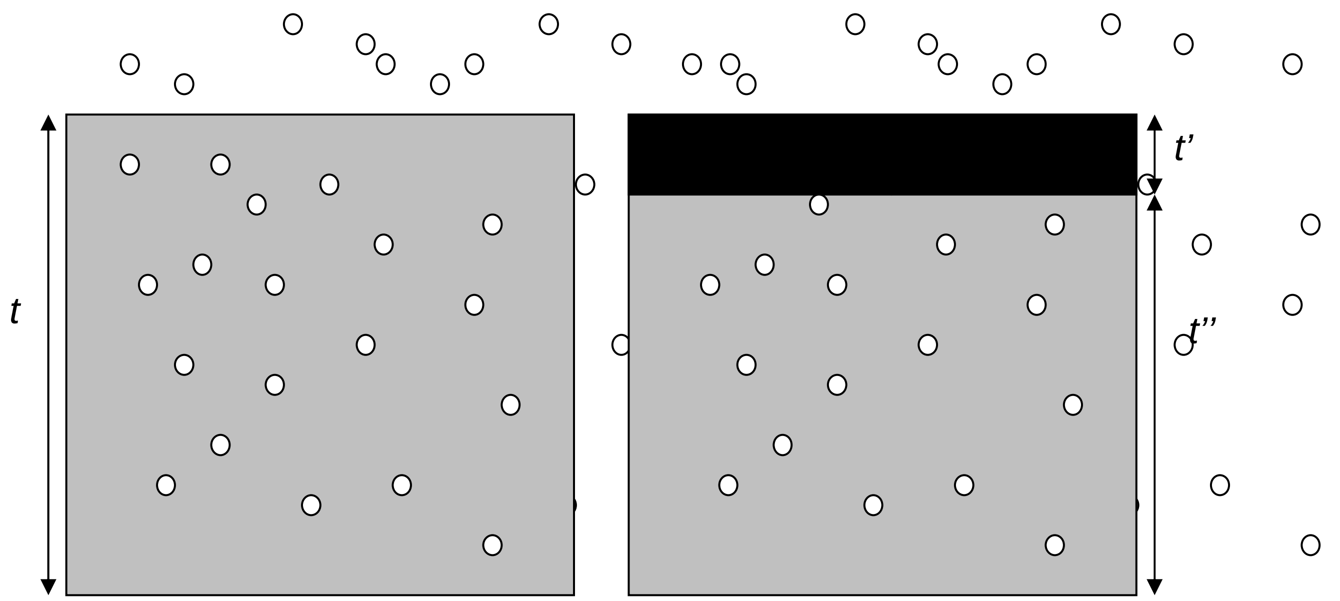
How to understand the variation of helium release with film thickness.

- **The helium is not being released from simple bulk diffusion.**
- **Instead, helium appears to only be originating from the near surface.**



Bulk Diffusion
ARF~α

2-Layer Diffusion
ARF~αt'/(t=t'+t'')

What I want you to take away from this presentation.

- What is the real ErH_2 film micro-structure.
  - Columnar grains
  - Texture-(111) out-of-plane
  - Oxide inclusions
  - Small oxide particulates
- How the helium behaves in the film.
  - Lenticular bubbles
  - Denuded zone
  - Constant helium release from near the film surface.